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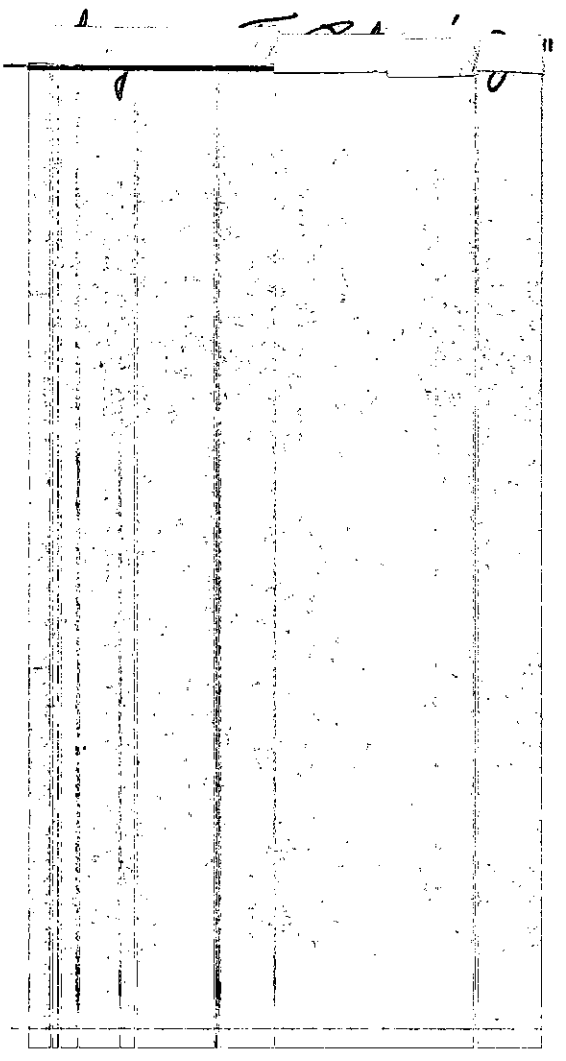


FLUORESCENCE YIELDS MEASUREMENTS

A THESIS

Presented to

the Faculty of the Graduate Division

Georgia Institute of Technology

In Partial Fulfillment

of the Requirements for the Degree

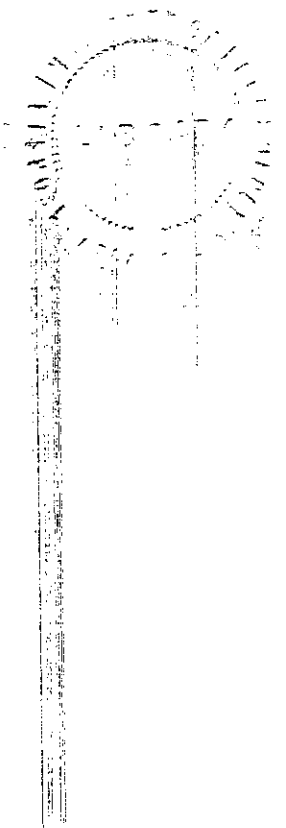
Doctor of Philosophy in the

School of Physics

By

Eugene Thayer Patronis, Jr.

June 1957



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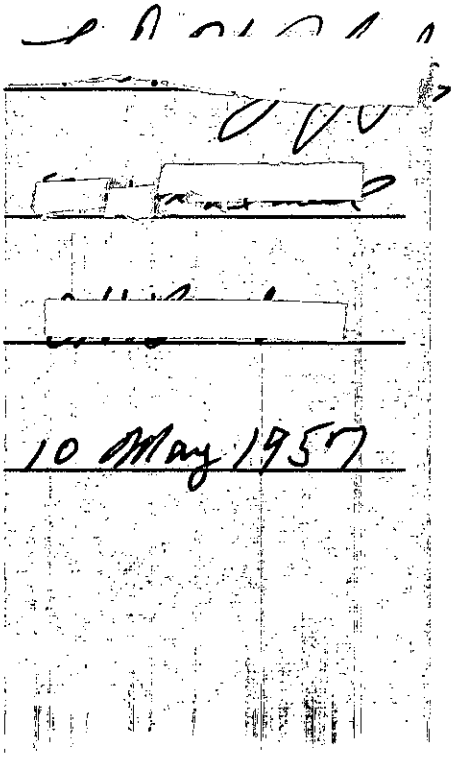

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ABSTRACT

Atomic fluorescence yields are important quantities in both atomic and nuclear physics. The fluorescence yield of a given atomic shell is defined as the reciprocal of the ratio of the number of atoms ionized in the given atomic shell to the number of these atoms which reorganize with the emission of an x-ray quantum appropriate to the given shell. The experimental determination of fluorescence yields is a problem which has occupied investigators since about 1925. Many ingenious methods have been evolved for fluorescence yields measurements. However, an analysis of these various experimental techniques indicates that many were either of very restricted applicability or required calculations of dubious validity in obtaining results from the raw experimental data. The primary difficulties of the early experimental work occurred because of the poor instrumentation available at the time the experiments were performed. Modern instruments should be able to overcome these faults and in addition should lead to experimental methods of sufficient scope so that measurements can be successfully performed on all elements of the periodic table.

The purpose of the present work is to develop a new experimental technique for making fluorescence yields measurements. The decision on the quality of an experimental method is based on the following three criteria: (1) Does the method have wide applicability, i.e., can measurements be made on a large number of elements? (2) Can results be obtained from the experimental data without recourse to theoretical calculations of questionable accuracy? (3) Is the method feasible from an economic standpoint?

In order to satisfy the first criterion, it is necessary for the fluorescence radiation detector to be sensitive throughout most of the x-ray spectrum. If, at the same time, the detector's response discriminates with regard to x-ray energy, the second criterion will be satisfied in large measure. It is found that of the modern radiation detectors such as photographic emulsions, cloud chambers, ionization chambers, Geiger counters, proportional counters, and scintillation counters, the gas-filled proportional counter has more of the desirable properties. A gas-filled proportional counter is chosen as a radiation detector in the present work for this reason.

The proportional counter requires a vacuum and gas system and an accurately regulated high voltage supply. These components were designed and constructed. Electronic amplification is provided for the pulses from the proportional counter by a fast linear amplifier of commercial design. In order to analyze the proportional counter output, a pulse height analyzer and scaler of commercial design are employed.

The source of x-ray excitation radiation is equally as important as the radiation detector. The interpretation of the experimental data is greatly simplified if a source of radiation of constant intensity and a simple known spectrum is employed. A long-lived radioisotope which decays by orbital electron capture has the desirable attributes and is chosen for a source of excitation radiation in the present work.

Before any serious experimental work can be undertaken it is necessary to ascertain the performance of the proportional counter and its associated electronic equipment. A sensitive measure of a proportional counter's performance is its energy resolution. It is possible to compute the expected resolution of a proportional counter from theory. The

energy resolution of the proportional counter used in the present work was measured experimentally using the radioisotope Fe^{55} and a comparison of the measured and theoretical values of the energy resolution indicates that the counter's performance is satisfactory.

In determining directly the fluorescence yield of a given atomic shell in a sample material it is necessary to know the number of atoms of the material which are ionized in the given shell and the number of these atoms which reorganize with the emission of an x-ray quantum appropriate to the given shell. In the present work the quantity of ionization produced in the sample material in the atomic shell of interest is determined from a measurement of the intensity of the excitation radiation to which the sample is exposed coupled with the known x-ray absorption properties of the sample material. Similarly, a measurement of the fluorescence radiation emitted by the sample coupled with the known x-ray absorption properties of the sample allows the determination of the number of atoms which reorganize with the emission of x-rays. In making the measurements of the intensities of excitation and fluorescence radiations with the proportional counter, a knowledge of the proportional counter's detection efficiencies for the excitation and fluorescence radiations is required. The efficiency of the counter for the detection of the excitation radiation was calculated using the known x-ray absorption properties of the proportional counter gas-filling mixture. The efficiency of the counter for the detection of the fluorescence radiation was experimentally determined by observing the fluorescence radiation spectrum as a function of counting gas pressure. It was possible to increase the counting gas pressure until practically all the fluorescence radiation entering the counter from the sample material was absorbed in the counting gas. This

method was not feasible in the case of the excitation radiation because of its higher energy.

The experimental method of the present work was employed to measure the K-shell fluorescence yields of the elements nickel, copper, and zinc, and the mean L-shell fluorescence yield of lead. The values obtained are for nickel, 0.33 ± 0.01; copper, 0.39 ± 0.01; zinc, 0.44 ± 0.01; and lead, 0.39 ± 0.01.

It is recommended that the present work be extended to include measurements of the K-shell fluorescence yields of other elements. Furthermore, it would be particularly desirable to make further L-shell fluorescence yields measurements because experimental values for these quantities are sparse. A worthwhile modification of the present method would be to use an x-ray machine in conjunction with a crystal monochromator as a source of excitation radiation. This modification would furnish a source of excitation radiation of variable energy and hence make possible the selective excitation of the L-subshells. In this way the fluorescence yields of the L-subshells could be obtained.

An examination of the theoretical calculations of fluorescence yields which have been made by various investigators indicates that there exists qualitative agreement between theory and the present experimental results. However, the fluorescence yields of only a few elements have been calculated. Fluorescence yields calculations present a worthwhile theoretical problem and modern computing techniques should make these calculations feasible.

CHAPTER I

INTRODUCTION

A quantity of considerable interest in atomic theory is the fluorescence yield. A knowledge of this quantity is useful in the interpretation of atomic spectra and in determining atomic wave functions. The K- and L-shell fluorescence yields are of value also in the study of nuclear decay involving orbital electron capture. The measurement of the K-shell yield has occupied investigators since 1925 while little work was done on L-shell measurements until 1935. Even though work in this field has continued until the present time, measurements on all elements have not been made, and many of the reported values are in doubt since they were obtained using techniques of questionable accuracy. Indeed, this is evidenced by the wide discrepancy which exists between values reported by different investigators. In particular, there is a necessity for a modern measurement of the L-shell fluorescence yields since it is among the reported values for the L-shell that the greatest discrepancies exist. It is the purpose of the present work to establish a somewhat different and more modern technique for measuring the K- and L-shell fluorescence yields. A report of the present work has been published in the Physical Review (1).

CHAPTER II

THE NATURE OF THE PROBLEM

The Phenomenon of Fluorescence.--Atomic fluorescence or the atomic fluorescence effect is the name given to the process wherein an atom which has been ionized in an inner shell has the inner shell vacancy filled by a higher shell electron with the subsequent release from the atom of a quantum of radiation whose energy is equal to the difference between the inner and higher shell binding energies. If the inner shell involved were the K-shell, the quantum of radiation would be characteristic of the K-series x-rays, either K_α, K_β, etc., depending on the higher shell involved; and the process would be termed K-shell fluorescence.

This is not the only manner in which an ionized atom may reorganize to become neutral. Another process which competes with atomic fluorescence is the Auger effect. In the Auger effect, instead of a quantum of radiation being released from the reorganizing atom, an electron which is referred to as an Auger electron is emitted from the atom. This Auger electron must always originate from a higher shell than the originally ionized shell in order for this type of reorganization to be energetically possible. The Auger effect is often referred to as a radiationless reorganization and was first interpreted by P. Auger (2).

When a vacancy in a given shell of an ionized atom is filled by a higher shell electron, the reorganization occurs either by means of

atomic fluorescence or the Auger effect. The fluorescence yield or the Auger yield for a given atomic shell is the probability of the occurrence of the respective process in the filling of the vacancy in that shell. For a macroscopic sample of an element, if N_k is the number of atoms ionized in the K-shell and n_k is the number of these atoms which reorganize by the emission of a K series quantum then ω_k, the K-shell fluorescence yield, is defined as

$$\omega_k = \frac{n_k}{N_k} . \qquad (1)$$

Similar definitions apply for shells other than the K-shell. Since either atomic fluorescence or the Auger effect account for all reorganizations, the Auger yield is given by

$$A_k = 1 - \omega_k . \qquad (2)$$

History of Fluorescence Yield Measurements.--The first serious work on the measurement of fluorescence yields was reported in 1925 by P. Auger (2). Auger used as a detector the then newly developed Wilson cloud chamber. Auger irradiated an atmosphere of argon in the chamber with x-rays of more than enough energy to produce K-shell ionization of the argon. The expansion photographs which Auger obtained showed the tracks of the argon K-shell photoelectrons and, in many events, demonstrated double tracks with a common origin. There was a marked difference in the lengths of the tracks forming the double track combinations. The length of the longer track was dependent on the energy of the initial ionizing radiation while the length of the shorter track was independent

of the energy of the initial radiation. Auger repeated his experiment with other gases and with different x-ray energies. He concluded that the shorter tracks were due to electrons which had been ejected by an energy transfer occurring during the reorganization of atoms which had been ionized in the K-shell. These electrons are now referred to as Auger electrons. Auger was able to estimate the K-shell fluorescence yields of the gases argon, krypton, and xenon by observing the abundance of the Auger electron tracks and the photoelectron tracks, the longer tracks, revealed in the expansion photographs of the Wilson cloud chamber. In addition, for the heavier gases krypton and xenon, Auger obtained estimates of the L-shell fluorescence yields in the same manner as before except that the energy of the incident radiation was lowered so that K-shell ionization could not occur. Auger's method, although direct, has the disadvantages that only materials in the gaseous state may be studied and a large number of photographs must be taken of the cloud chamber in order to obtain statistical accuracy.

A method of measuring fluorescence yields that has wider application than Auger's method is one which consists of measuring the x-ray power incident on, and fluorescent from, a secondary radiator formed of the material whose fluorescence yield is to be determined. In this method, a primary beam of x-rays from an x-ray tube produces fluorescence radiation in a radiator of high atomic weight. This fluorescence radiation, which consists of a few characteristic lines, is collimated into a narrow beam, and the power in this beam is measured by means of an ionization chamber. This collimated x-ray beam is then used to irradiate the sample under investigation and the power of the fluorescence

radiation from the sample is measured by a second ionization chamber. The fluorescence yield of the sample may be calculated from these two power measurements provided that the solid angle subtended by the second ionization chamber and the absorption coefficients, frequencies, and relative intensities of the x-ray lines in the incident and fluorescence beams are known. Many investigators have used this method or some variation of this method. In 1920 Harms (3) used this method in measuring ω_k, the K-shell fluorescence yield of Fe, Cu, Zn, Se, and Sr. In the work of Harms, the radiation incident on the secondary radiator was filtered in an attempt to render this radiation monochromatic. Harms' reported values for the fluorescence yields were in error because of an invalid computation of the x-ray power from the ionization chamber current readings. Balderston (4) used the same method in the measurements of ω_k for Fe, Ni, Cu, Zn, Mo, and Ag. The radiation incident on the secondary radiator in Balderston's work had been monochromatized by reflection from a crystal in a Bragg spectrometer. Compton (5) has pointed out that Balderston made serious errors in his computations by assuming that the ionization produced in air by x-rays of different energies is proportional to their absorption in air. Balderston's assumption is false since scattering makes an important contribution especially at higher x-ray energies. Martin (6) employed this method with a filtered beam of radiation incident on the secondary radiator to measure ω_k for Mo, Se, and Ni. In 1933 Stephenson (7) applied the same method for the determination of $\omega_{L_{III}}$ for uranium and later, in 1937, for the determination of ω_k for Ni, Cu, Zn, Se, Zr, Mo, Rh, Ag, Cd, and Sn.

In 1932 Locher (8) returned to Auger's method to measure ω_k for oxygen, neon, and argon. One-thousand-nine-hundred-and-fifty stereoscopic pictures were taken of tracks formed in a Wilson cloud chamber which had been irradiated with monochromatic x-radiation. Although the statistical accuracy of Locher's work was good, the identification of the Auger electron tracks was difficult, especially in the light gases oxygen and neon.

Haas (9) obtained data on ω_k for elements of low atomic number. In Haas' work, monochromatic incident radiation was obtained by reflection from a crystal mounted in a vacuum spectrometer. The fluorescence radiation was measured by an ionization chamber. Haas reported values of ω_k for the elements Mg, Si, S, Cl, Ca, and Cr.

In 1934 Lay (10) made measurements of ω_k in the region of atomic numbers from 16 to 34 using the x-ray power comparison method previously discussed. Lay, however, used a photographic emulsion technique rather than ionization chambers for the determination of the x-ray powers. Lay's results differed widely from those reported by other investigators probably because of inaccuracies arising in the evaluation of the darkening of the photographic emulsions.

In the years immediately preceding, during, and immediately after World War II, little or no work was done in the field of fluorescence measurements. When work was resumed new methods were introduced that had a number of advantages over the older techniques.

Kinsey (11), in 1948, introduced a method for determining the L-shell fluorescence yields for the radioisotopes ThB, ThC, and RaD. Geiger counters were used to detect both L-series x-rays and the alpha or beta particles given off by the radioactive sources. In Kinsey's

arrangement, one Geiger counter measured the number of L-series quanta emitted while another measured the number of decays by detecting alpha or beta particles. Kinsey deduced the L-shell fluorescence yields by comparing the number of events detected by the separate Geiger counters. In other work, Kinsey (12) was able to calculate L-shell fluorescence yields by a comparison of measured x-ray line widths. The values so determined do not agree well with the values obtained by direct means. However, Kinsey's work is often used to obtain estimates of fluorescence yields that have not been measured.

A method of measuring fluorescence yields of gaseous substances with greater accuracy than Auger's original method was introduced in 1949 by Curran (13). Curran used a proportional counter filled with the gas whose yield was to be determined. The counting gas was irradiated with monochromatic x-rays. Voltage pulses of two distinct amplitudes were obtained from the proportional counter. The larger voltage pulses correspond to the total expenditure of the x-ray quantum energy in the counting gas. The smaller voltage pulses occur when the atoms of the counting gas reorganize with the emission of K-series x-radiation which escapes from the counter. Curran determined the K-shell fluorescence yield of the gas by comparing the numbers of the larger and smaller voltage pulses. In 1950 West and Rothwell (14) extended these measurements to include gases of high atomic numbers.

In 1952 Broyles, Thomas, and Haynes (15) gave a comprehensive review of the work done on fluorescence yield measurements. These workers reported measured values of the Auger yields of Ba and Hg. These yields were determined by observing the relative intensities of Auger electron

lines and internal conversion electron lines for the radionuclides Cs^{137} and Au^{198}. The observations were made with a magnetic lens spectrometer employing a thin window Geiger counter.

In 1953 Roos (16) made a determination of ω_k for the elements Zr, Nb, Mo, Rh, Pd, Ag, Cd, and Sn. Roos irradiated target foils with monochromatic x-radiation which had been obtained by crystal reflection. The fluorescence radiation as well as the x-radiation incident on the target foils were measured with a scintillation spectrometer. Roos' measurements were extended in 1955 to include the elements Fe, Ni, Cu, and Zn (17).

In 1955 Harrison (18) using a proportional counter measured ω_k for argon by the method of Curran. This work also included a determination of ω_k for Cu and Y which was obtained by comparison of K x-ray and K Auger line intensities in the K capturing radioisotopes Zn^{65} and Sn^{113}.

In 1955 Gray (19) measured the K Auger yields of Y, Po, and U by comparing Auger line intensities and internal conversion line intensities using a magnetic lens beta ray spectrometer and the method of Broyles.

Choice of Method.--In choosing a method of measuring fluorescence yields for the present work, a thorough review of previously used methods was made with the view of determining the shortcomings inherent in these methods. The criteria used in evaluating different experimental methods of measuring fluorescence yields were the following:

1) Does the method have wide applicability, i.e., can measurements be made on a large number of elements?
2) Can results be obtained from the experimental data without recourse to theoretical calculations of questionable accuracy?

3) Is the method feasible from an economic standpoint?

Auger's cloud chamber method does not satisfy the first criterion listed above since it is applicable only to samples which are in the gaseous state at ordinary temperatures. The x-ray power comparison method as employed by Harms (3) and others required a large number of theoretical calculations to be made in order to convert ionization chamber current readings into determinations of x-ray power. The substitution of photographic plates for the ionization chambers by Lay (10) merely substituted one extended theoretical calculation for another and thus does not satisfy the second criterion. Kinsey's method (11) employing Geiger counters fails the first criterion since measurements can be made only on certain radioactive isotopes. Curran's method (13), though good, is limited to gaseous samples. The method of Broyles and his coworkers (15) using a magnetic lens spectrometer is only effective for elements at the upper end of the periodic table because of the difficulty encountered in detecting Auger electrons of energy less than about ten kev. The method of Roos (16) is not well adapted for working with elements of low atomic number because of the difficulty involved in detecting photons of energy less than about eight kev by the use of scintillation counters.

In order to satisfy the first criterion, it is necessary to use a detector which can operate efficiently over a range of x-ray energies from a few hundred ev to about 100 kev. This is the energy range encompassed by the atomic fluorescence radiation from about 80 per cent of the elements. At the same time, it is desirable that the detector

give a response which is proportional to the energy of the x-radiation being detected so that the detector can discriminate between radiations of different energies. This simplifies the analysis of the data which helps satisfy the second criterion. The gas-filled proportional counter chosen for a detector in the present work has the characteristics desired of a fluorescence radiation detector.

The source of x-radiation used to excite fluorescence radiation in a sample material should be monochromatic to further insure that the second criterion is satisfied. If the source of excitation is monochromatic or consists of only a few discrete energies, only known absorption coefficients and x-ray line intensities must be used in the calculation of the amount of ionization produced in a sample material. A suitable method of obtaining excitation radiation of constant intensity is to employ a relatively long-lived radioisotope which decays by pure K capture. Only the K-and L-series x-rays characteristic of the daughter atom will be present and the L-series x-rays, being of much lower energy, may be removed from the beam by means of an absorber. The excitation radiation might also be obtained either by filtering the radiation from an x-ray tube or reflecting the radiation from an x-ray tube by a crystal. In either case some provision for monitoring or maintaining a constant intensity of the x-radiation from the x-ray tube would have to be made.

The experimental method used in this work consists of irradiating the sample under investigation with x-radiation furnished by a relatively long-lived radioisotope which decays by K capture and measuring the number of incident and fluorescence quanta by means of a proportional counter. The analysis of the experimental data obtained in this way

does not necessitate doubtful calculations as will be indicated more fully in Chapter V.

It is only fair to point out some of the shortcomings of this method. First, the K-shell fluorescence yields of the elements with atomic numbers greater than 75 cannot be measured by this method since the x-ray energy available from relatively long-lived radioisotopes which decay by K capture is insufficient to produce K-shell ionization in these elements. For elements of Z greater than 75 an x-ray machine would have to be used as the source of excitation energy. Second, the accuracy inherent in the method is poor if the Auger yield of the sample of interest is very small due to the fact that the error is of the same magnitude as the Auger yield in the region of low Auger yields. The method of Broyles which employs the detection of the Auger Electrons in determining the Auger yield would be more suitable in this case. The L-shell fluorescence yields of all elements with Z greater than about 20 lie in the range where the detection of the L-shell quanta could lead to more accurate results than the detection of the corresponding Auger electrons.

CHAPTER III

APPARATUS

A block diagram of the apparatus used in the present work appears in Fig. 1. The chief components of the system are 1) proportional counter, 2) counting tube power supply, 3) amplifier, 4) pulse height analyzer, 5) scaler, and 6) source of excitation. A description of the individual components is given below.

Proportional Counter.--A description of the proportional counter can best be given by describing its two constituents, the counting tube and vacuum system, separately.

Figure 2 is a cross-section view of the counting tube. The counting tube is a cylinder made of Alcoa grade 2SF aluminum. This grade of aluminum was chosen because it is practically free of elements other than aluminum, hence fluorescence radiation from the counter wall is widely separated in energy from the fluorescence radiation from the samples of interest. The cylinder has a length of 30 centimeters, an inside diameter of four centimeters, and a wall thickness of 0.4 centimeter. The ends of the cylinder are closed off by threaded lucite plugs. The anode of the counter is a five mil tungsten wire (Sylvania Electric Company process NS 30) placed coaxially within the aluminum cylinder. The anode is held in position by Kovar seals mounted in the center of each of the lucite end plugs. Two ports are provided for the counting tube. These two ports are separated by 90 degrees as measured on the axis of the cylinder and are both 15 centimeters from the end of the

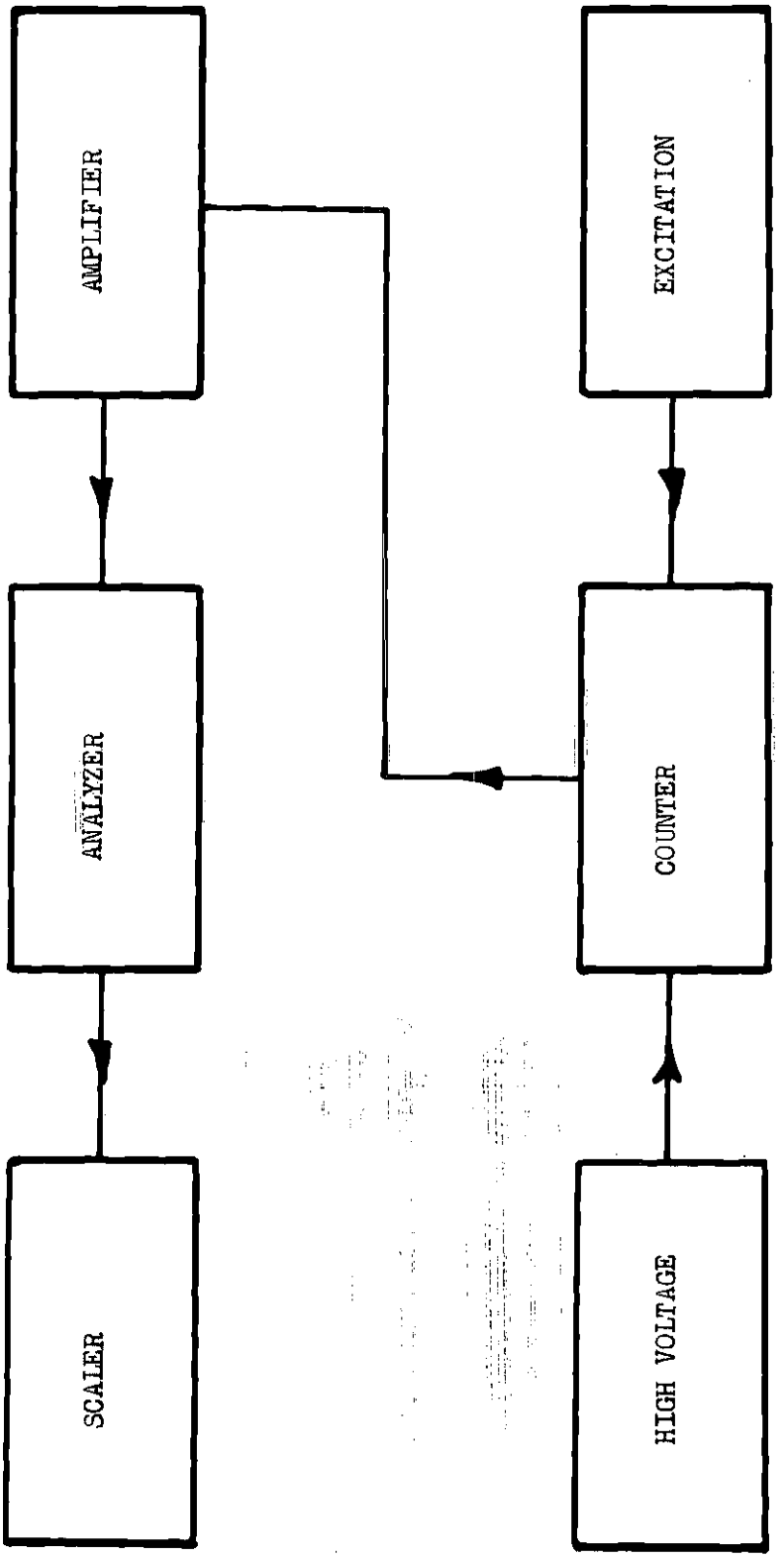
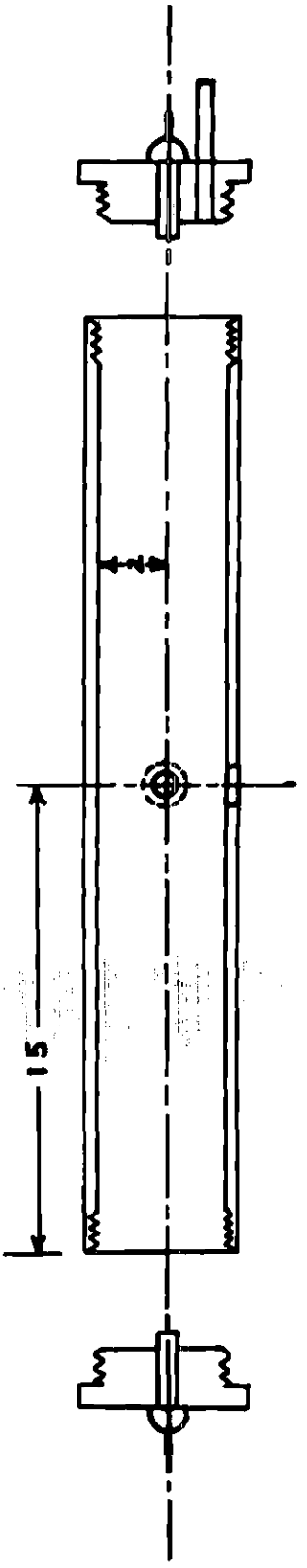


Fig. 1. Block Diagram of Experimental Apparatus



NOTE: DIMENSIONS IN CENTIMETERS

Fig. 2. Construction Details of the Proportional Counting Tube

counting tube. Each port was made by drilling a one-fourth inch hole through the cylinder wall and then countersinking. One of the ports is fitted with an aluminum foil window of 6.62 mg per cm^2 thickness. The window of the second port is formed from the sample material under investigation. The counting tube is connected to the vacuum system by means of a three-fourth inch aluminum pipe which is screwed through a hole in one of the lucite end plugs. The other end of the aluminum pipe terminates in a type 304 Hoke valve. All vacuum seals were made with a mixture of beeswax and rosin recommended by John Strong (20).

Figure 3 is a schematic drawing of the vacuum system. The vacuum system was constructed from copper and brass tubing. All joints in the system were made either by hard soldering or by flange connections using rubber "O" ring-type seals. The valves used in the system are of the gate type which have been modified by the method of Kurie (21) in order to make them effective under vacuum conditions. Provisions are made for the attachment of three gas bottles to the system. The pressure in the vacuum system is measured with an Ashcroft type 1004 Bourdon gauge and an NRC type 05-0100 thermocouple gauge. The vacuum system employs an NRC Model H-2 oil diffusion pump and a Welch Duo-Seal mechanical pump. The counting gas employed in the proportional counter is a mixture of 92 per cent argon and eight per cent carbon dioxide ($1\!4$) which was supplied by the Ohio Chemical and Surgical Equipment Company. The impurity content of the gas is guaranteed to be less than 0.5 per cent.

The energy resolution of the counter was measured with the aid of the associated electronic apparatus described later in this chapter and the radioisotope Fe^{55} as a source. Fe^{55} decays by K capture to Mn^{55}

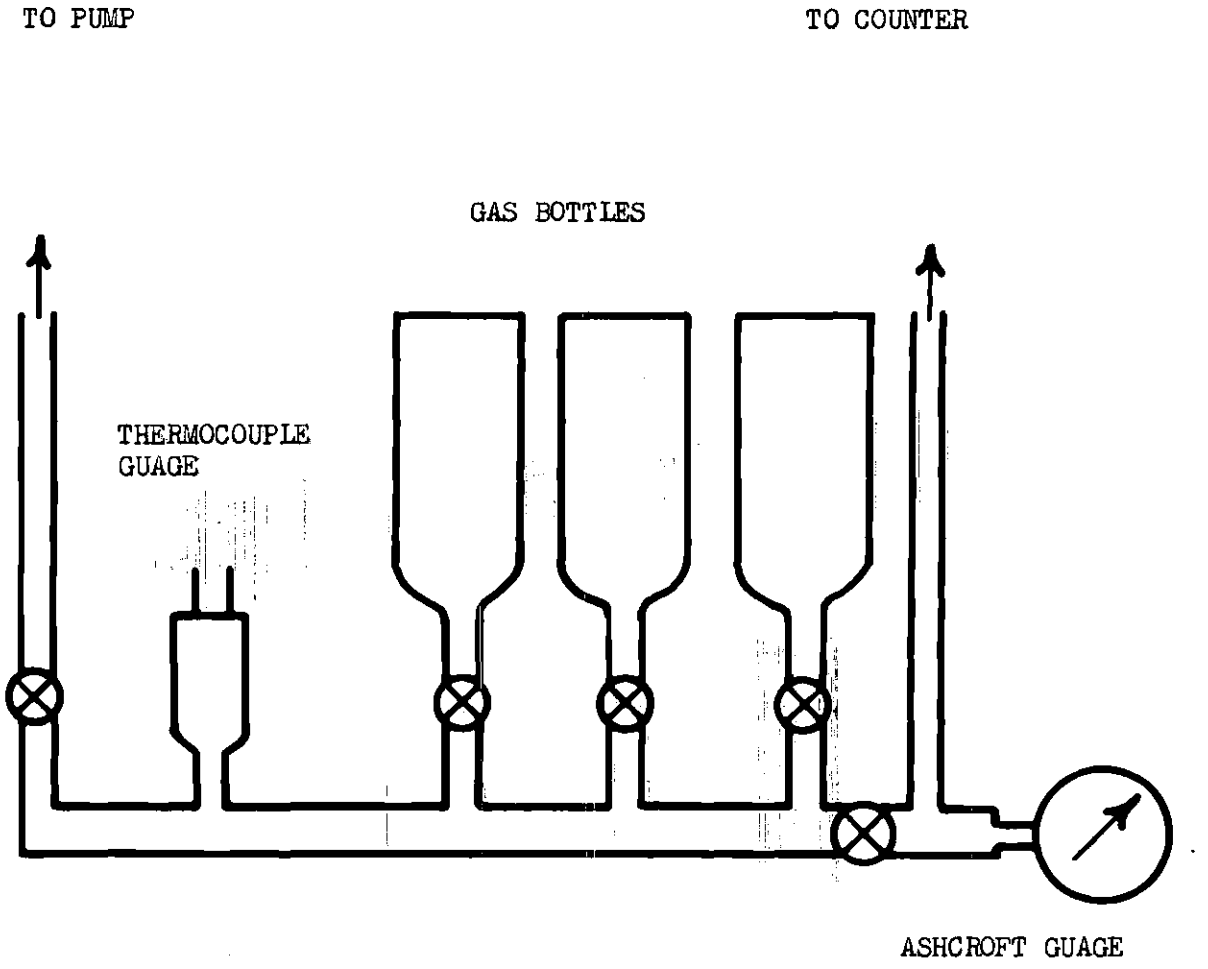


Fig. 3. Schematic Drawing of Vacuum System

with the emission of the K-series x-rays of the daughter atom. Figure 4 is a plot of the pulse counting rate versus pulse height obtained with the counter upon examination of the Fe^{55} photon spectrum. The larger peak represents the K-series x-radiation from Mn and corresponds to an energy of 5.9 kev. The smaller peak is the A escape peak mentioned previously and corresponds to an energy of 3.0 kev. The energy resolution of the counter is defined as the width of the peak at half its maximum ordinate divided by the abscissa of the peak position. The energy resolution for the Mn K radiation was found to be 0.21. In order to evaluate the counter's performance it is necessary to compare the measured resolution with the resolution which is theoretically obtainable from a proportional counter under the conditions of operation employed.

The spread in pulse size obtained from a proportional counter when the counter is observing a monochromatic source of radiation is due principally to two causes. First, there are random fluctuations in the amount of initial ionization produced in the counting gas by the passage of an ionizing radiation through the gas and second, there are random fluctuations of the gas multiplication about its average value. Curran (13) has shown that these two effects contribute in about equal measure to the total spread of counter pulse size. Snyder (22) has made a theoretical calculation of the spread in pulse size produced by a proportional counter. Snyder found that

$$(\Delta H)^2 = \frac{2\,(\bar{H})^2}{\bar{n}} \qquad (3)$$

where $(\Delta H)^2$ is the mean square deviation in the size of the pulse, $\bar{H}$ is the average pulse size, and $\bar{n}$ is the average number of ion pairs produced

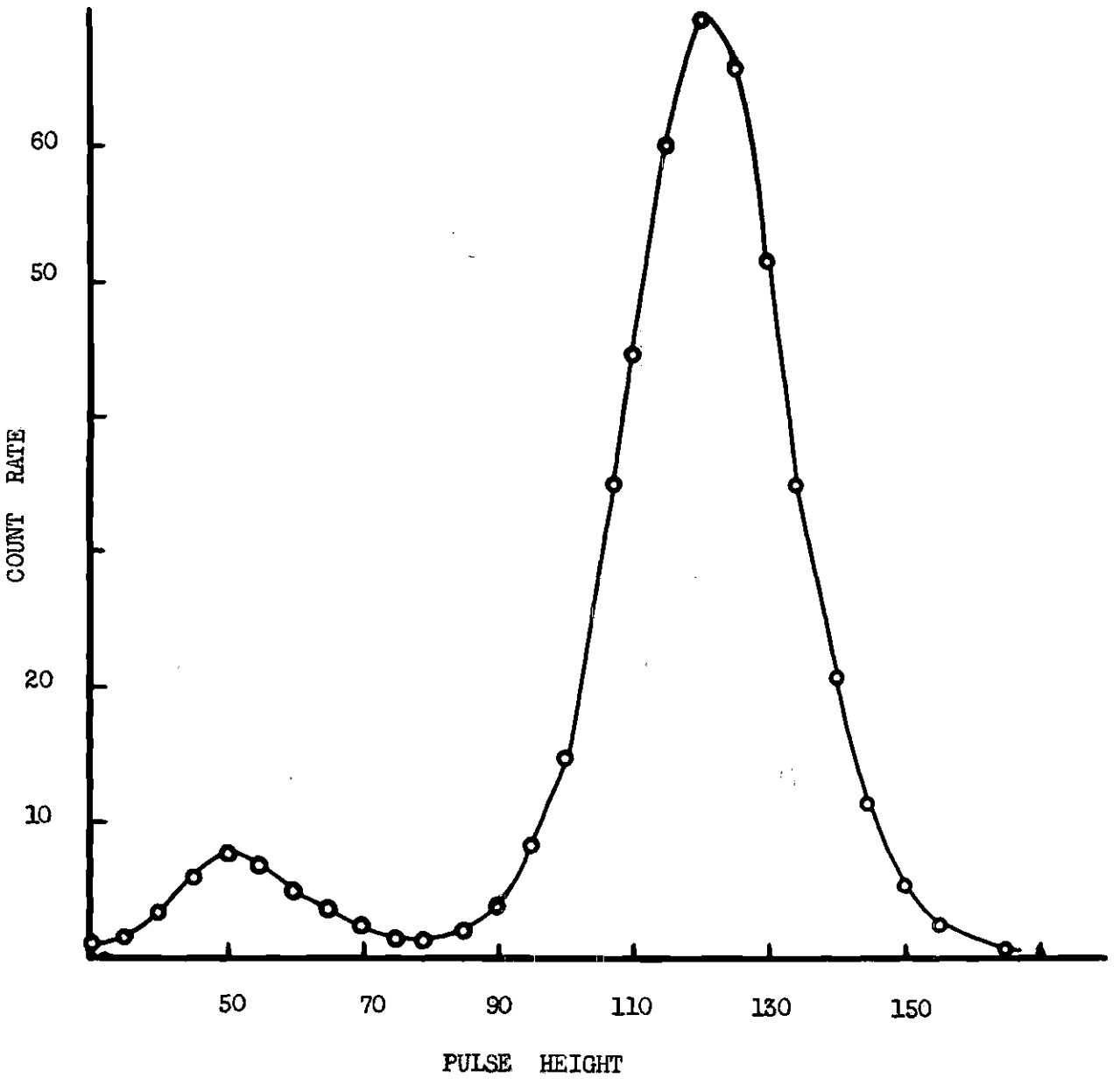


Fig. 4. Spectrum of Fe^{55} Obtained with Proportional Counter

in the counter by the primary ionizing radiation. The resolution as previously defined is approximately twice the root mean square deviation of pulse size divided by the average pulse size (23), so that

$$\frac{2(\Delta H)}{\bar{H}} = \frac{2\sqrt{2}}{(\bar{n})^{1/2}} . \tag{4}$$

The average energy required to produce an ion pair in the counter gas is 30 ev (24). The energy of the x-radiation from the Fe^{55} source is 5.9 kev so that $\bar{n}$ is given by

$$\bar{n} = \frac{5.9 \times 10^3}{30} = 1.97 \times 10^2 . \tag{5}$$

The theoretical resolution is then found to be, using equations (4) and (5),

$$\frac{2\,\Delta H}{\bar{H}} = \frac{2\sqrt{2}}{(1.97 \times 10^2)^{1/2}} = 0.202 . \tag{6}$$

The measured value of the energy resolution is 0.21, indicating that the counter operation is satisfactory.

Counting Tube Power Supply.--A regulated high voltage power supply was designed and constructed. The circuit diagram is given in Fig. 5. The two to three kilovolts required to operate the proportional counting tube is obtained from this circuit with a short term regulation of 0.01 per cent and a long term regulation of 0.1 per cent. In this circuit, the radio frequency output of a tuned plate oscillator is fed into a conventional half-wave rectifier by means of a step-up transformer. The output voltage of the power supply is regulated by controlling the screen

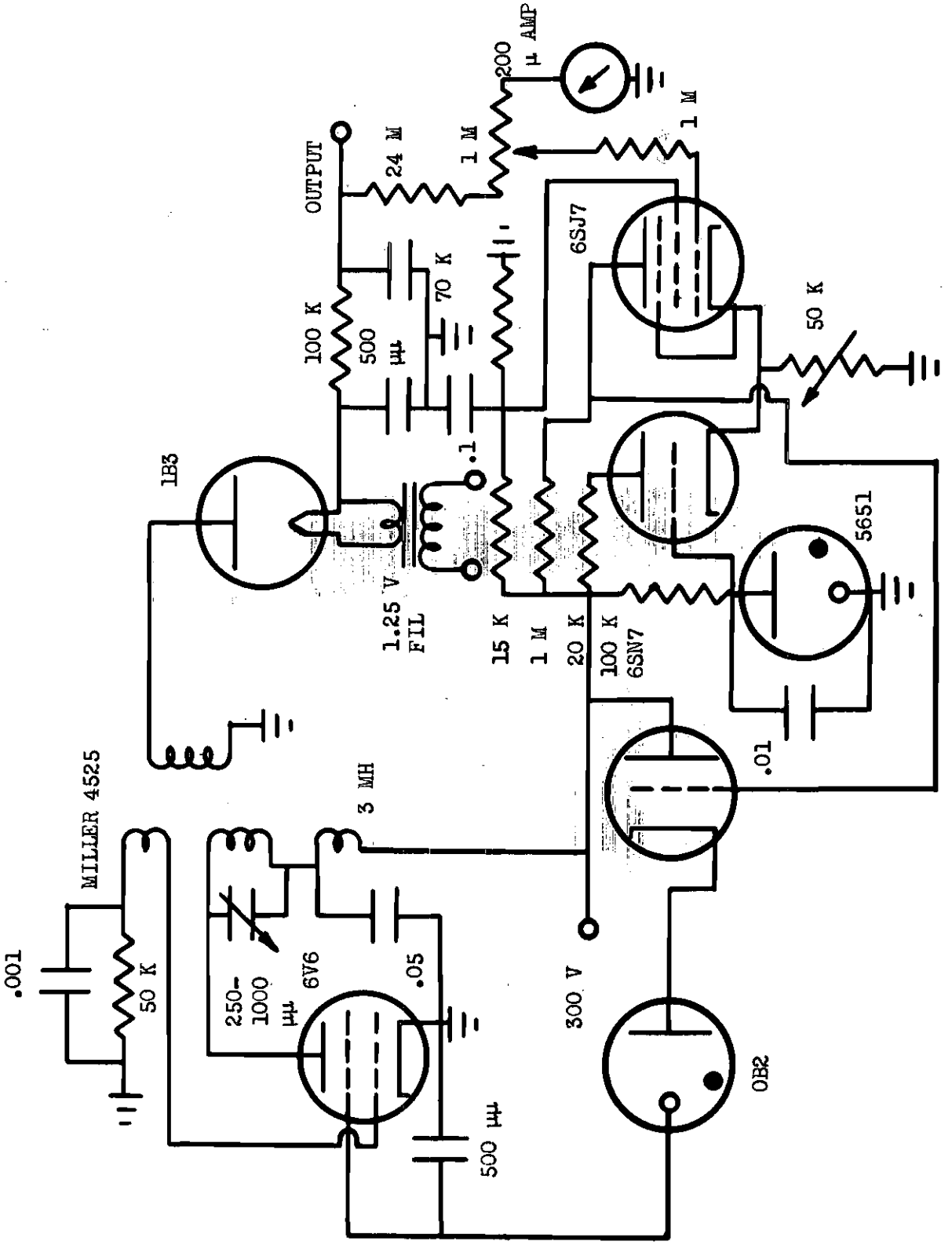


Fig. 5. Circuit Diagram of Proportional Counter High Voltage Supply

voltage on the oscillator tube which determines the output of the oscillator.

Amplifier.--The linear pulse amplifier used in the present work is an Atomic Instrument Company Model 204B. The noise and overload characteristics of the amplifier were improved by the insertion of crystal diodes in the grid circuits of the stages where the pulse polarity is independent of the input pulse polarity. This modification did not appreciably increase the rise time of the amplifier above its rated value of 0.2 microseconds.

Pulse Height Analyzer.--Pulse height analysis was performed with a Radiation Counter Laboratory Model 2204 single channel differential pulse height analyzer. This analyzer accepts pulses with amplitudes between zero and 80 volts with a window width which is variable from zero to about eight volts.

Scaler.--Pulse rates were determined with an Atomic Instrument Company Model 1020A scaler. This scaler has a time resolution of five microseconds and a maximum scale of 256. The counting times were measured with a Standard Electric Time Company ten second sweep clock.

Source of Excitation.--The source of excitation radiation in the present work was the radioisotope Cd^{109}. Cd^{109} decays by K and L orbital electron capture to Ag^{109} with the emission of K- and L-series x-radiation characteristic of the daughter atom. A nuclear gamma ray is also emitted as a result of the decay since the transition does not lead directly to the ground state of the Ag nucleus. This nuclear gamma ray has an energy of 89 kev and the absorption of this gamma ray in the materials investigated was negligible. The L x-radiation was almost totally absorbed in

the source itself. As a result, all ionization in the sample materials was due to the K-series x-radiation from Ag which has an energy of about 22 kev. The source was evaporated from a solution of CdCl onto a lucite screw. The details of the source mounting and the source collimator are shown in Fig. 6. The source collimator was constructed from concentric cylinders of lead and aluminum. The source was held in position over the ports of the counting tube by means of a bakelite ring slipped over the outside wall of the counting tube. A hole was drilled and tapped in the bakelite ring for the admission of the collimated source. One end of the collimator seated directly against the port over which the collimator was positioned.

In addition to the foregoing apparatus it was found convenient to employ a fast oscilloscope to visually examine the amplified electrical pulses from the proportional counter and a pulse generator to check the performance of the electrical circuits. The oscilloscope employed was a Tektronix type 514D. The pulse generator used was designed by the author and has been described in Nucleonics (25).

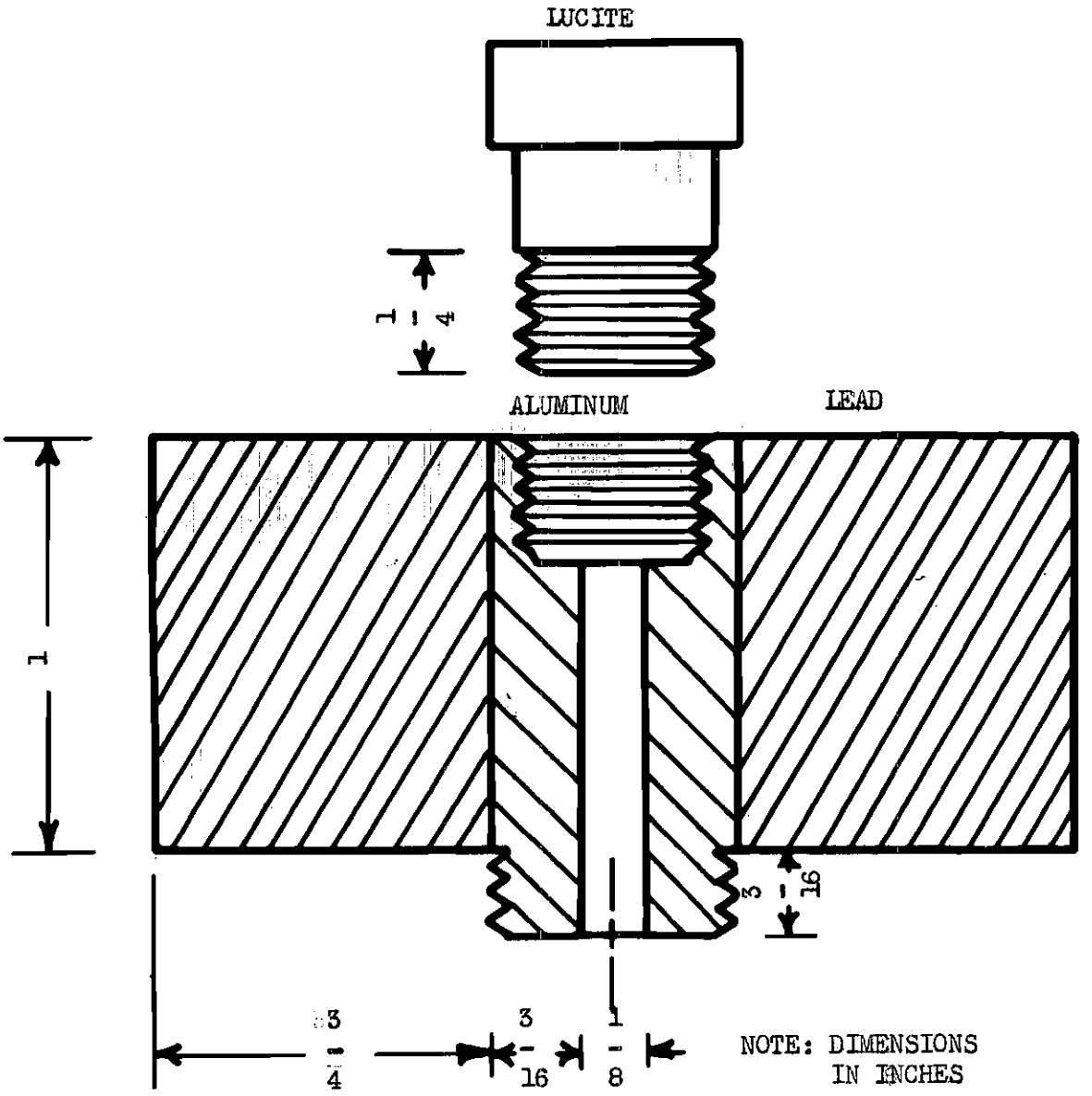


Fig. 6. Construction Details of Source Collimator

CHAPTER IV

EXPERIMENTAL PROCEDURE

The materials investigated were obtained in the form of foils of thickness ranging from one to five mils. The thicknesses of the individual samples were determined by weighing a known area of the sample with a Christian Becker Chainomatic beam balance and then calculating the thickness from the measured weight and the known density of the sample. After the thickness of a sample had been determined, a window was prepared from the sample, and the window was installed in one of the ports of the counting tube. The counter side of the window was covered with a 15 mg per cm^2 polythene foil to absorb Auger electrons. The counting tube was then attached to the vacuum system and evacuated. The counter was pumped on for a period of two days before it was filled with the gas mixture of argon and carbon dioxide to an absolute pressure of about one atmosphere. All electronic equipment was turned on at the beginning of the evacuation period so that it would have ample time to reach thermal equilibrium before the counter was ready for active use.

After the counter had been filled with the gas, the collimated source was positioned over the port with the aluminum window and a pulse height analysis of the counter signals was performed. The collimated source was then positioned over the port containing the sample under investigation and a pulse height analysis was again performed. This procedure, along with a measurement of the background in the counter, was repeated several times. Finally, the collimated source was again

positioned over the port containing the sample under investigation and a pulse height analysis of the signals from the counter was made using different counting gas pressures in the counter. The counter operating pressure was varied from 0.5 atmosphere to 1.75 atmospheres.

CHAPTER V

ANALYSIS OF DATA

Figure 7 illustrates the geometry of the source of excitation, the sample material, and the counting tube. In general, the radiation from the source will contain several discrete x-ray lines with energies E_i, i being 1, 2, 3,....l, where l is the total number of lines. The photoelectric absorption coefficient of the sample material for an x-ray line of energy E_i is μ_i. The intensity at the sample of an x-ray line with energy E_i is n_i. Let dI_k be the number of atoms ionized in the K-shell in an axial element of the sample of thickness dx located at the position x. Then dI_k is given by

$$dI_k = P_k \sum_i \mu_i \quad n_i \exp(- \mu_i \ [t-x]) \, dx, \tag{7}$$

where P_k is the probability that the photoelectric absorption occurs in the K-shell. P_k may be found from

$$P_k = 1 - R_k \tag{8}$$

where R_k is the ratio of the x-ray absorption coefficients on the low and high frequency sides of the K absorption edge (26).

The number of atoms ionized in the K-shell that reorganize with the emission of K-series fluorescence radiation is

$$\omega_k dI_k \tag{9}$$

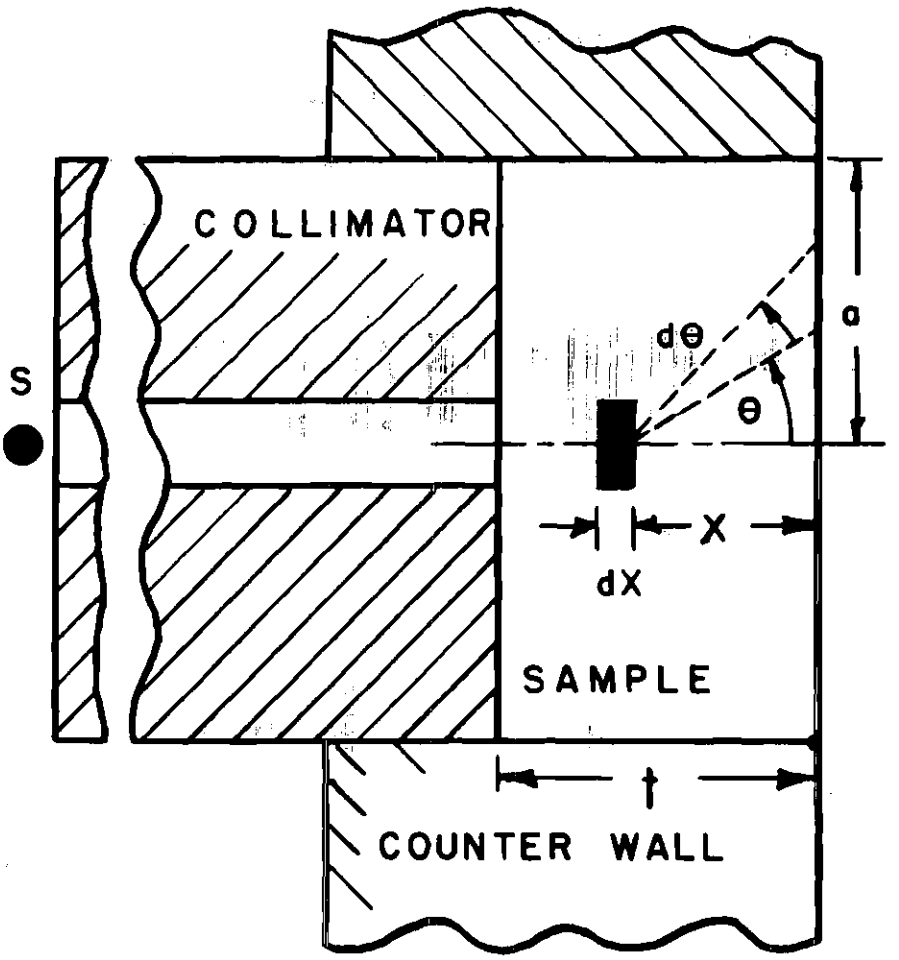


Fig. 7. Geometry of Source of Excitation, Sample Material, and Counting Tube

where ω_k is the K-shell fluorescence yield. The K-series fluorescence radiation will be isotropic (27) and will consist of several discrete lines with energies E_f and intensities N_f, where f = 1, 2, 3,....m, m being the number of K-series fluorescence x-ray lines. The intensities are normalized so that $\sum_f N_f = 1$. The absorption coefficient of the sample for a fluorescence x-ray line of energy E_f is μ_f. The amount of fluorescence radiation emitted by the axial element of the sample which enters the counting tube is then

$$\frac{1}{2}\,\omega_k\,dI_k \int_0^{\tan^{-1}\frac{a}{x}} \sum_f N_f \exp(-\mu_f\, x \sec\theta)\,\sin\theta\, d\theta\,. \quad (10)$$

Finally, the total amount of fluorescence radiation entering the counting tube from all the axial elements of the sample is

$$F = \frac{1}{2} P_k \omega_k \int_{x=0}^{t} \int_{\theta=0}^{\tan^{-1}\frac{a}{x}} \sum_i \mu_i n_i \exp(-\mu_i [t-x]) \sum_f N_f \exp(-\mu_f\, x \sec\theta)$$

$$\sin\theta\, d\theta\, dx\,. \quad (11)$$

Although the calculation of F could be carried out in its exact form, it is convenient to make some simplifying approximations. The K-series radiation from the source of excitation is composed almost entirely of the lines K_{α_1}, K_{α_2}, and K_{β_1}, with intensities of 0.575, 0.282, and 0.138, respectively (28). The respective energies of these lines as

determined from Hill's tables (29) are 22.160 kev, 21.988 kev, and 24.942 kev. These x-ray energies are nearly the same and the absorption coefficients of the sample materials investigated vary smoothly with energy in the energy range of these lines, hence, a reasonable approximation is to use a single effective absorption coefficient which corresponds to the weighted average energy of the excitation x-ray lines. This same approximation can be made in determining an effective absorption coefficient for the fluorescence radiation from the sample material. With these approximations, the sums appearing in the expression for F are reduced to a single term each and

$$F = 1/2\, P_k \omega_k \bar{\mu}_i N_i \int_{x=0}^{t} \int_{\theta=0}^{\tan^{-1}\frac{a}{x}} \exp\left[-\bar{\mu}_i (t-x)\right] \exp(-\bar{\mu}_f x \sec\theta) \sin\theta \, d\theta \, dx \qquad (12)$$

where $\bar{\mu}_i$ is the effective absorption coefficient for the excitation radiation, $\bar{\mu}_f$ is the effective absorption coefficient for the fluorescence radiation, and N_i is the intensity of the excitation radiation at the sample surface, i.e., $N_i = \sum_i n_i$. Note that N_f has disappeared from the expression for F because $\sum_f N_f = 1$.

The evaluation of the various factors appearing in equation (12) is now discussed. The thickness, t, of the sample material is known from the measurement described in Chapter IV. The energies of all x-ray lines are determined from the tables of Hill (29). The x-ray absorption

coefficients are obtained from the tabulated data in the Handbook of Chemistry and Physics (30). The relative intensities of the x-ray lines are taken from Williams (28). The integral appearing in the expression for F may now be evaluated graphically.

In order to determine F, the amount of fluorescence radiation entering the counter, a graph of the pulse counting rate versus the pulse height for the fluorescence radiation detected by the counter was made. The area of this graph is F multiplied by the efficiency of the counter for the fluorescence radiation. The efficiency of the counter was determined by studying the areas of a sequence of these graphs as a function of the gas pressure in the counting tube. Evidently, the area of such a graph is proportional to the counter efficiency at that pressure. It was possible to increase the pressure in the counting tube until practically all the fluorescence radiation entering the counting tube was absorbed, i.e., until the efficiency of the counter for the fluorescence radiation was nearly unity. A graph of counter efficiency versus counter pressure is presented in Fig. 8.

In order to determine N_i, the intensity of the x-ray beam incident on the sample, a graph of the pulse counting rate versus pulse height for the excitation radiation detected by the counter was also made. The area of this graph is N_i multiplied by the efficiency of the counter for the excitation radiation provided a small correction is made for the absorption which occurs in the thin aluminum window of the counting tube. The efficiency of the counter for the excitation radiation could not be determined as it was for the fluorescence radiation because it was not feasible to increase the pressure in the counter until practically all of the excitation photons were absorbed. The efficiency was studied as a function

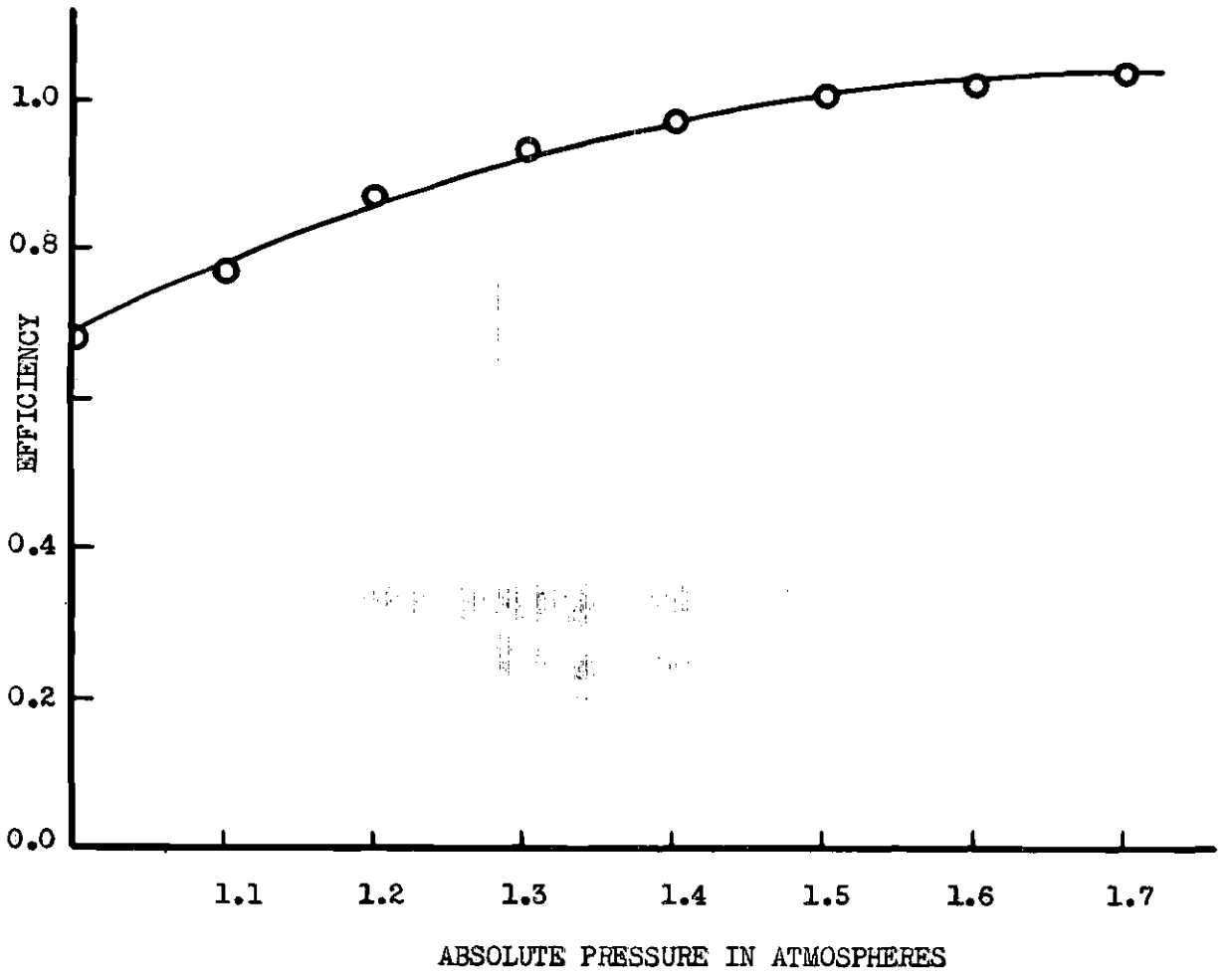


Fig. 8. Curve of Efficiency versus Pressure Obtained with a Nickel Sample

of pressure, however, to verify the absence of any wall effect. In contrast to the fluorescence radiation, the excitation radiation is a collimated x-ray beam and, therefore, has a well defined absorption path in the counting tube equal to the inside diameter of the counting tube. The absorption coefficient of the argon gas for the excitation radiation can be evaluated using tabulated absorption data (30). The efficiency of the counter for the excitation radiation is, thus, simply $1-\exp(-\mu_c d)$ where μ_c is the absorption coefficient of the gas for the excitation radiation and d is the inside diameter of the counting tube.

In terms of the experimental quantities,

$$F = \frac{A}{Q_f} \tag{13}$$

where A is the area of the graph of pulse counting rate versus pulse height for the fluorescence radiation and Q_f is the efficiency of the counter for the fluorescence radiation and,

$$N_i = \frac{B}{Q_i} \exp(\mu_w t_w) \tag{14}$$

where B is the area of the graph of the pulse counting rate versus pulse height for the excitation radiation, and μ_w and t_w are, respectively, the absorption coefficient and thickness of the aluminum window.

Finally,

$$\omega_k = \frac{2\,\frac{A}{Q_f}}{\frac{B}{Q_i}\exp(\mu_w t_w) P_k \mu_i I} \tag{15}$$

where

$$I = \int_{x=0}^{t} \int_{\theta=0}^{\tan^{-1}\frac{a}{x}} \exp[-\bar{\mu}_i(t-x)] \exp(-\bar{\mu}_f x \sec\theta) \sin\theta \, d\theta \, dx \, . \qquad (16)$$

Thus, ω_k can be calculated once the factors $F = \frac{A}{Q_f}$ and $N_i = \frac{B}{Q_i}$ are determined from the experimental data.

The L-shell fluorescence yield, ω_L, is found by use of expressions similar to (15) and (16) except that P_k is replaced by P_L, the probability that photoelectric absorption occurs in the L-shell. The factor P_L may be calculated from tabulated data on x-ray absorption coefficients (30) by noting that

$$P_L = 1 - R_{I,\,II} R_{II,\,III} R_{III,\,M} \qquad (17)$$

where $R_{I,\,II}$ is the ratio of the x-ray absorption coefficients on the low and high frequency sides of the L_I absorption edge, $R_{II,\,III}$ is the ratio of the x-ray absorption coefficients on the low and high frequency sides of the L_{II} absorption edge, and $R_{III,\,M}$ is the ratio of the x-ray absorption coefficients on the low and high frequency sides of the L_{III} absorption edge (27).

CHAPTER VI

RESULTS AND DISCUSSION

Table 1 lists the effective absorption coefficients, sample thicknesses, probabilities of K- or L-shell ionization, and the values of the integral I equation (16). Table 2 lists the areas, A and B, of the graphs of counting rate versus pulse amplitude, the counter efficiencies Q_i and Q_f, and the correction factor for absorption of excitation radiation in the aluminum counter window.

The areas of all graphs were determined using a K and E compensating polar planimeter. Each area was measured with the planimeter at least ten times and the error associated with the measured value of an area was taken to be the greatest observed deviation from the arithmetic mean of the ten measurements. This error was found to be approximately one per cent. The error associated with the efficiency of the counter for the excitation radiation is about two per cent. This is due to an uncertainty about the gas pressure in the counter and corresponds to an error in the pressure of approximately ten per cent. The errors in the quantities entering into the calculations of the fluorescence yields are then one per cent in A, one per cent in B, one per cent in Q_f, two per cent in Q_i, and 1.5 per cent in I since the evaluation of the integral depends on the product of two measured areas. The probable error in the fluorescence yields is then $\pm$ three per cent. Table 3 lists the observed fluorescence yields along with their probable errors.

Table 1. Experimental Quantities

Atomic Number	$\bar{\mu}_{i^{-1}}$ cm	$\bar{\mu}_{f^{-1}}$ cm	t cmx10^{-3}	P_k	P_L	I
28	214	525	1.045	0.880		4.81×10^{-4}
29	231	453	4.94	0.879		4.64×10^{-4}
30	221	365	13.5	0.875		1.15×10^{-4}
82	851	908	5.02		0.754	2.07×10^{-5}

Table 2. Graphical Areas and Counter Efficiencies

Atomic Number	Area A	Area B	Q_i	Q_f	Window Correction
28	8.52	32.68	0.042	0.69	1.017
29	9.76	31.30	0.042	0.69	1.017
30	3.05	31.34	0.042	0.82	1.017
82	1.10	34.90	0.042	0.50	1.017

Table 3. Measured Values of Fluorescence Yields

Atomic Number	ω_K	ω_L Averaged Over All Sub-shells
28	0.33 ± 0.01	
29	0.39 ± 0.01	
30	0.44 ± 0.01	
82		0.39 ± 0.01

A comparison of the results of the present work with those of other investigators indicates that the K-shell values are in good agreement with the measurements of Arends in all cases and with Berkey in the case of Zn while the other values listed by Burhop tend to be slightly higher (26). Recent measurements by Harrison (18) on Cu and Roos (17) on Ni, Cu, and Zn are in agreement with the present results. The L-shell result for Pb is in good agreement with the measurements for this region of the periodic table made by Lay (10) and Kinsey (11) but is higher than Kinsey's estimates based on x-ray line widths.

Even though measurements were made on just a few elements, the method is adaptable for measurements of the K- and L-shell fluorescence yields for about 80 per cent of the elements. In particular, it would be desirable to adapt the method to measure the fluorescence yields of the various sub-shells of the L-shell. This could be done by using an x-ray machine and a crystal monochromator as a source of excitation radiation. In this way, the sub-shells of the L-shell, L_I, L_{II}, and L_{III}, could be selectively excited and ω_{L_I}, $\omega_{L_{II}}$, $\omega_{L_{III}}$ could be individually determined. These fluorescence yields would be of value in nuclear problems involving internal conversion and K and L orbital electron capture.

CHAPTER VII

THEORETICAL CONSIDERATIONS

The theory of the Auger effect was first given in 1927 by Wentzel (31), with subsequent contributions to the theory being made by Møller in 1931 (32). Theoretical calculations of the fluorescence yield have been made by Burhop (33), Pincherle (34), Burhop and Massey (35), and Rubenstein and Snyder (36). The following discussion is based on the recent survey of theoretical methods given by Burhop (26).

In the Auger effect, the system of interest consists of an atom which in the initial state is ionized in an inner level. The final state of the system consists of the atom with an inner level occupied and with one electron, an Auger electron, in a state of positive energy i.e., in a free particle state. According to the method of Wentzel the perturbation inducing an Auger transition is the direct coulomb interaction between the two electrons involved in the radiationless transition of the atomic system from the initial to the final state. The Auger transition probability i.e., the number of Auger processes occurring per second involving the atomic states i and f, is

$$b_n = 2\pi\hbar^{-1}\left|f_1^n - f_2^n\right|^2 \tag{18}$$

where f_1^n is given by

$$\iint \psi_f^*(\bar{r}_1,\bar{r}_2)\,\frac{e^2}{|\bar{r}_1-\bar{r}_2|}\,\psi_i(\bar{r}_1,\bar{r}_2)\,d\bar{r}_1 d\bar{r}_2 \tag{19}$$

and f_2^n is given by

$$\iint \psi_f^*(\bar{r}_2,\bar{r}_1)\,\frac{e^2}{|\bar{r}_1-\bar{r}_2|}\,\psi_i(\bar{r}_1,\bar{r}_2)\,d\bar{r}_1 d\bar{r}_2 \ . \qquad (20)$$

In these expressions, b_n is the nonrelativistic radiationless transition rate, $\frac{e^2}{\bar{r}_1-\bar{r}_2}$ is the direct coulomb interaction operator between electrons located at $\bar{r}_1$ and $\bar{r}_2$, respectively, ψ_i and ψ_f are the wave functions describing the two electrons involved in the transition in the initial and final states, respectively. For example, ψ_i might represent an electron in the L-shell and an electron in the M-shell when there is a vacancy in the K-shell and ψ_f would represent an electron which has occupied the vacancy in the K-shell and an electron which has been ejected from the atom, an Auger electron. In these expressions, the final state wave function ψ_f is normalized to represent one ejected electron per unit time per unit energy range. The term $f_1^n - f_2^n$ appears in equation (18) in order to take into account the fact that the initial and final state wave functions must be antisymmetrical in the coordinates of the two electrons. The properly symmetrized form of the initial state wave function would then be

$$2^{-1/2}\left\{\psi_i(\bar{r}_1,\bar{r}_2) - \psi_i(\bar{r}_2,\bar{r}_1)\right\} \qquad (21)$$

and similarly for the final state wave function. Following this method of Wentzel, Burhop has made theoretical calculations of the fluorescence yield of the K-shell in silver (33). In making his calculations, Burhop used screened hydrogenic wave functions.

Further theoretical calculations have been made by Massey and Burhop (35) according to a relativistic theory. Massey and Burhop used four component Dirac wave functions and the method of treating the perturbations due to the interaction of two electrons as introduced by Møller. This calculation involves the consideration of a two-step process in which the vacancy in the inner shell is filled through a radiative transition. The radiation field for this transition is computed. In the second step of the process, a higher shell electron is considered to be ejected through the interaction with this radiation field. In the relativistic calculations, the Auger transition rate is given by

$$b_r = 2\pi\hbar^{-1} \left| f_1^r - f_2^r \right|^2 \tag{22}$$

where f_1^r is given by

$$\int \Psi_f^*(\bar{r}) \left\{ -ea_o - e\boldsymbol{\alpha}\cdot\bar{a} \right\} \Psi_{ia}(\bar{r}) d\bar{r} \tag{23}$$

and f_2^r is given by

$$\Psi_f^*(\bar{r}) \left\{ -ea_o - e\boldsymbol{\alpha}\cdot\bar{a} \right\} \Psi_{ib}(\bar{r}) d\bar{r} \ . \tag{24}$$

In these expressions, Ψ_{ia} and Ψ_{ib} are Dirac wave functions describing the Auger electron in its possible initial bound states and Ψ_f is the Dirac wave function describing the Auger electron in its final free particle state, $\boldsymbol{\alpha}$ is the usual matrix associated with the Dirac equation, and the term $\left\{ -ea_o - e\boldsymbol{\alpha}\cdot\bar{a} \right.$ is the operator representing the interaction between an electron and a radiation field (37) in which

$$a_o(\bar{r}) = \int \frac{1}{|\bar{r}-\bar{r}^1|} \exp(2\pi i \nu_{fi} \frac{|\bar{r}-\bar{r}^1|}{c}) \rho_{fi}(\bar{r}^1) d\bar{r}^1 \qquad (25)$$

$$\bar{a}(\bar{r}) = c^{-1} \int \frac{1}{|\bar{r}-\bar{r}^1|} \exp(2\pi i \nu_{fi} \frac{|\bar{r}-\bar{r}^1|}{c}) \bar{j}_{fi}(\bar{r}^1) d\bar{r}^1 \ . \qquad (26)$$

Physically, $a_o(\bar{r})$ represents the retarded scalar potential due to a charge distribution described by ρ_{fi} and $\bar{a}(\bar{r})$ represents the retarded vector potential due to a current density described by $\bar{j}_{fi}$. The charge and current distributions are considered to arise from the electron which undergoes a radiative transition i.e., it emits a radiation field which is then absorbed by the Auger electron leading to the Auger electron's ejection from the atom.

$$\rho_{fi} = -e \chi_f^* \chi_i \qquad (27)$$

$$\bar{j}_{fi} = e \chi_f^* \alpha \chi_i \qquad (28)$$

χ_i and χ_f are the Dirac wave functions describing the initial and final states of the electron which undergoes the radiative transition. It is to be noted that the Dirac wave functions employed describe individual electrons only and cannot be made up of properly symmetrized wave functions describing two electrons. In the equation for b_r, the difference $f_1^r - f_2^r$ is taken so as to make the relativistic expressions correspond to the nonrelativistic expressions at the nonrelativistic limit.

The fluorescence yield of a given atomic level is the ratio of the total radiative rate feeding that level to the sum of the total Auger rate and radiative rate feeding that level from all other possible atomic levels. The fluorescence yield for the p level is then given by

$$\omega_p = \frac{\sum_n a_n^p}{\sum_n \sum_m b_{n\,m}^p + \sum_n a_n^p} \tag{29}$$

where $\sum_n a_n^p$ is the summation of radiative transition rates over all levels of energy greater than the p level and $\sum_n \sum_m b_{nm}^p$ is the summation of Auger transition rates over all pairs of levels for which an Auger transition to the p level is energetically possible. In the calculations of the a's, the radiative transition rates, only electric dipole transitions need be considered. The a's are then determined by equations of the form

$$a_n^p = 64\,\pi^4\,\nu_{np}^3 \left|\frac{M_{np}}{3hC}\right|^2 \tag{30}$$

where ν_{np} is the frequency of the radiation emitted and is equal to $\frac{E_n - E_p}{h}$ and M_{np} is the matrix element of the electric dipole moment which is given by

$$M = \int \chi_f^* e\bar{r}\, \chi_i d\bar{r} \tag{31}$$

χ_f is the wave function describing an electron in the p level and χ_i is the wave function describing the electron in the n level. The dipole moment operator is $e\bar{r}$.

The theoretical calculations of Auger, Burhop, and Massey and Burhop have led to the conclusions that the Auger transition rate is essentially independent of atomic number whereas the radiative transition rate is proportional to the fourth power of the atomic number. It follows

then from the equation for ω_p that the fluorescence yield should be of the form $\frac{\alpha Z^4}{\beta + \alpha Z^4}$ where α and β are constants. This may be written as $(1 + \gamma Z^{-4})^{-1}$ where $\gamma = \frac{\alpha}{\beta}$. Thus the theoretical expression for the fluorescence yield is

$$\omega = (1 + \gamma Z^{-4})^{-1} . \qquad (32)$$

Burhop (26) has made a comparison of the theoretical prediction, equation (32), with the average values of the K-shell fluorescence yields as reported by investigators prior to 1952. Burhop found good agreement using as γ the value 1.12×10^6. Figure 9 is a graph of the K-shell values obtained in the present work presented in the form of $\frac{\omega_k}{1-\omega_k}$ versus Z^4. The straight line obtained corresponds to a γ of 1.14×10^6.

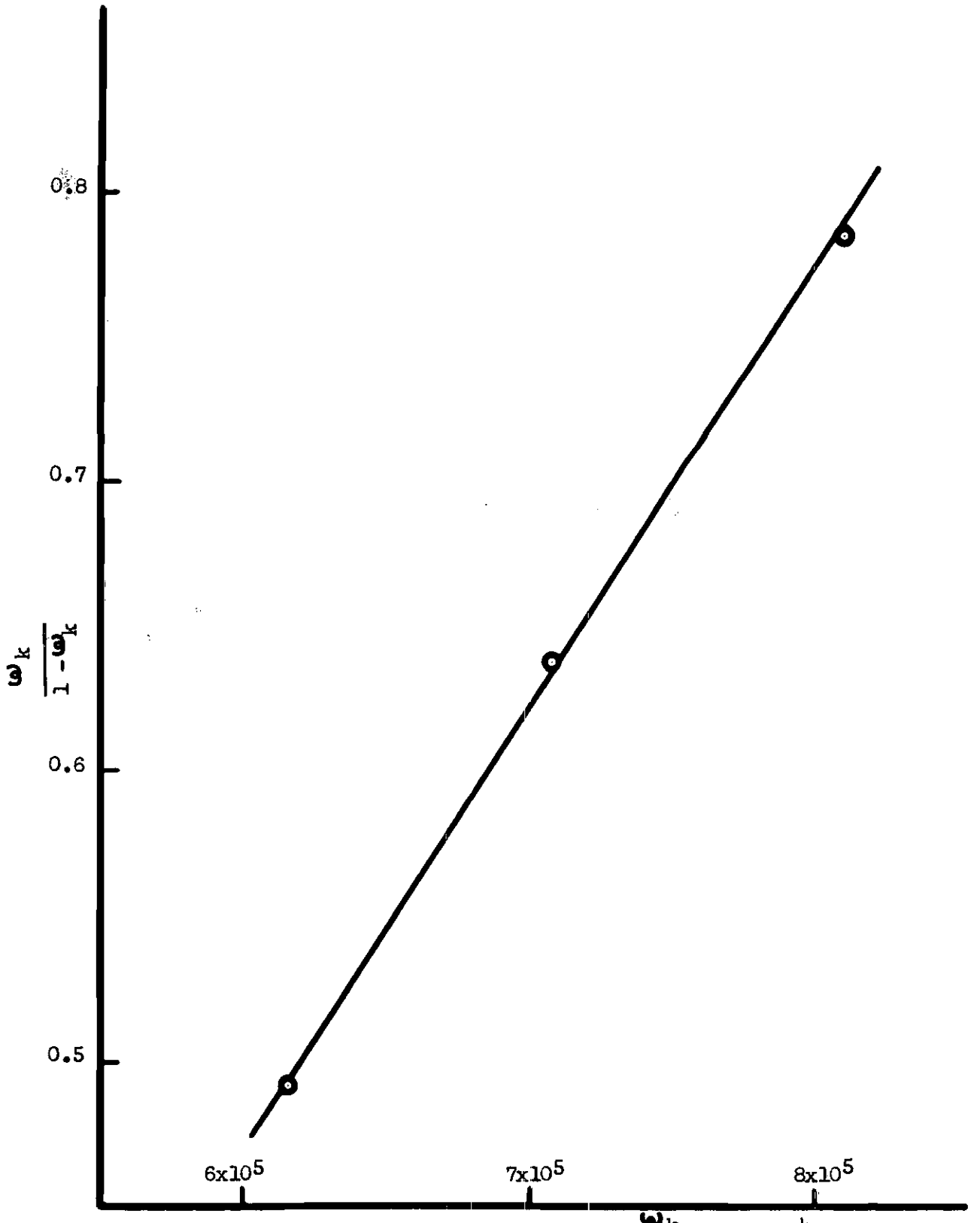


Fig. 9. Measured Variation of $\frac{\omega_k}{1-\omega_k}$ with Z^4

APPENDIX

SAMPLE CALCULATION

Figure 10 is a graph of the pulse counting rate versus pulse height obtained with a nickel sample. The area included under the 7.5 kev peak is the area A of equation (13). This area as listed in Table 2 is 8.52. The efficiency of the counter for the nickel fluorescence radiation from Table 2 is 0.69. From equation (13),

$$F = \frac{A}{Q_f}, \qquad F = \frac{8.52}{0.69} = 12.34 . \tag{13}$$

From equation (14),

$$N_i = \frac{B}{Q_i} \exp(\mu_w t_w) . \tag{14}$$

The area B from Table 2 is 32.68, Q_i is 0.042, and the window correction $\exp(\mu_w t_w)$ is 1.017 so that N_i becomes

$$N_i = \frac{(32.68)(1.017)}{(0.042)} = 792 .$$

From equation (15),

$$\omega_k = \frac{2\,\frac{A}{Q_f}}{\frac{B}{Q_i} \exp(\mu_w t_w) P_k \bar{\mu}_i I} .$$

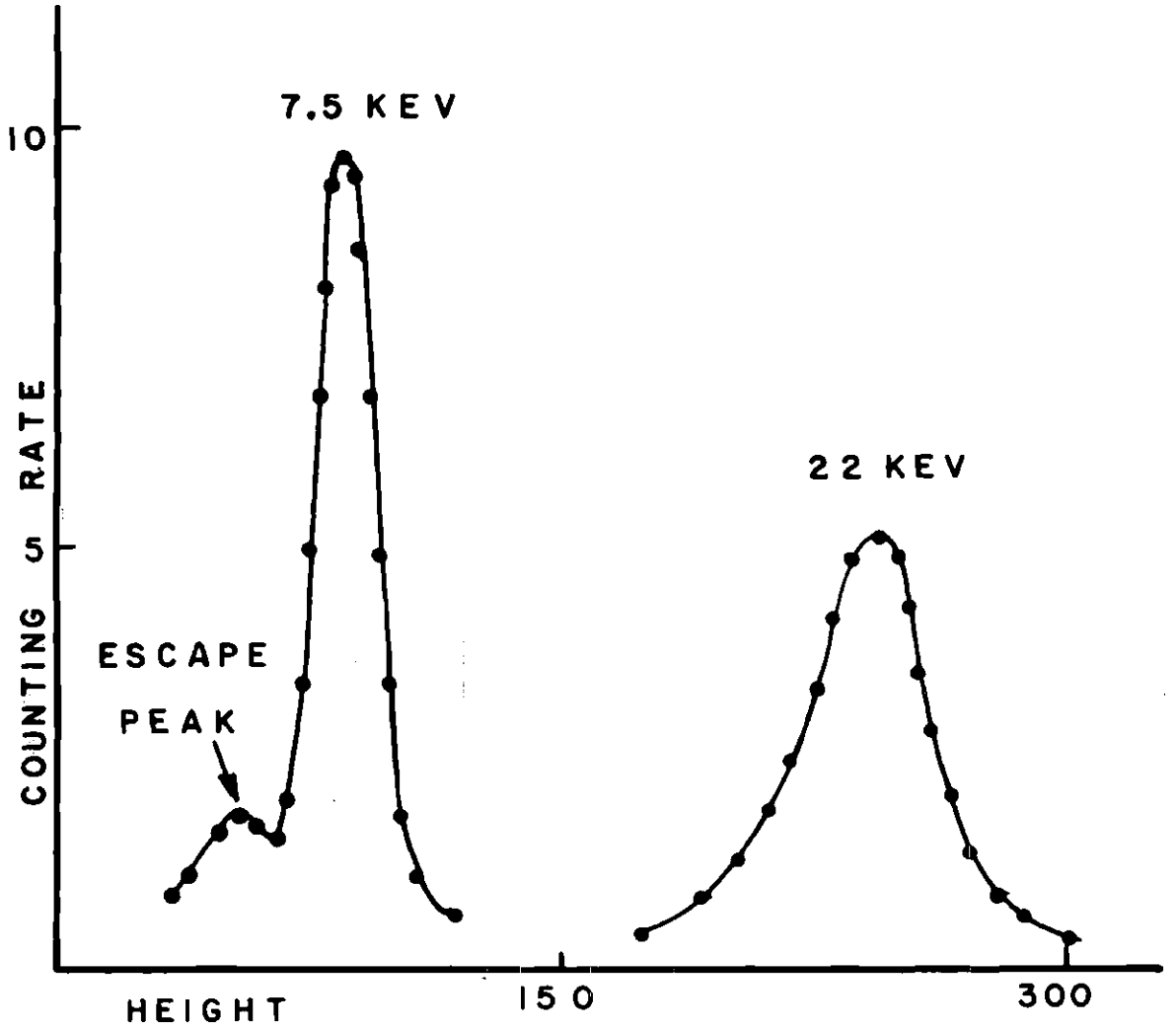


Fig. 10. Fluorescence and Excitation Spectra Obtained with a Nickel Sample

From Table 1, P_k is 0.880, $\bar{\mu}_i$ is 214, and I is 4.81×10^{-4}. Therefore,

$$\omega_k = \frac{(2)(12.34)}{(792)(0.880)(214)(4.81 \times 10^{-4})} = 0.33 .$$

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